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Characterization of dc magnetron sputtered indium oxide films

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Abstract : Indium Oxide films were deposited by dc reactive magnetron sputtering from indium target on to glass substrates held at temperatures in the range 373–673 K in an oxygen partial pressure of 2×10^{-4} mbar. The dependence of structure, electrical and optical properties of the films on the substrate temperature was systematically studied The films prepared at 373 K were amorphous while those formed at temperatures ≥ 473 K were polycrystalline with cubic structure The electrical resistivity of the films decreased from $2.4 \times 10^{-2}\ \Omega$ cm to $1.3 \times 10^{-3}\ \Omega$ cm and Hall mobility increased from 2.5 cm^2/Vsec to 18 cm^2/Vsec with the increase of substrate temperature from 373 K to 673 K due to the improvement in the crystallinity of the films The temperature dependence of Hall mobility indicated that the grain boundary scattering of the charge carriers predominated in these films The optical transmittance (at λ = 600 nm) increased from 68% to 85% and the optical band gap increased from 3.64 eV to 3.78 eV with the increase of the substrate temperature from 373 K to 673 K respectively.

Keywords . Indium oxide films, dc magnetron sputtering, electrical and optical properties

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1. Introduction

In recent years, there is an increasing interest in the development of transparent conducting thin film materials due to their importance in a wide range of applications such as solar cells, liquid crystal displays and photoconductors. One of such materials is Indium oxide. Indium oxide (In_2O_3) films with high electrical conductivity and high transmittance in the visible range were prepared by various deposition techniques such as thermal oxidation of indium [1,2], reactive evaporation [3-6], pulsed laser deposition [7], dc magnetron sputtering [8-10], rf sputtering [11] and spray pyrolysis [12]. In recent years, much attention is focussed on films prepared by dc reactive magnetron sputtering, because of the advantage of sputtering from indium target with high deposition rates and uniformity over large area substrates by easily controlling the composition of the films. The physical properties of the films prepared by dc reactive magnetron sputtering are highly influenced by the sputtering parameters such as oxygen partial pressure, substrate temperature and sputtering pressure. In this investigation, we made an attempt to deposit indium oxide films by dc reactive magnetron sputtering at different substrate temperatures and characterized then by studying their structural, electrical and optical properties.

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2. Experimental

Indium oxide films were deposited onto ultrasonically cleaned glass substrates by dc reactive magnetron sputtering from a home made planar magnetron sputtering system [13] capable of obtaining an ultimate vacuum of 2×10^{-6} mbar. A circular planar magnetron of 100 mm diameter was used as the magnetron cathode. The magnetron was designed in such way that two permanent ring magnets made of samarium cobalt are cooled by water circulation [14]. The magnetron target assembly was mounted on the top plate of the vacuum chamber so that the sputtering can be performed in sputter down mode. A continuous variable dc power supply of 750 V and 3 A was used as a power source. The sputter target was pure indium of 100 mm diameter and 3 mm thick. 99.995% pure argon and oxygen were used as sputtering and reactive gases respectively. The flow rates of both the gases were individually controlled by Tylan mass flow controllers. Indium oxide films were deposited under various substrate temperature in the range 373 – 673 K in an oxygen partial pressure of 2×10^{-4} mbar and sputtering pressure of 4×10^{-2} mbar. Before deposition of each film, the indium target was pre-sputtered in pure argon for 15 minutes in order to remove the surface oxide layer formed if any, on the target. The following characterization measurements were carried out at 303 K on the as-deposited indium oxide films. The thickness of the films was measured using interference method. The crystallographic structure of the films was analyzed with X-ray diffraction using copper radiation with a wavelength of 0.15406 nm. The electrical resistivity and Hall mobility of the films were measured employing standard van der Pauw method. The optical transmittance and reflectance of the films were recorded using Hitachi double beam spectrophotometer.

3. Results

The indium oxide films deposited by dc reactive magnetron sputtering were pin hole free and highly adherent to the surface of the substrate. The thickness of the films investigated was in the range 80 - 120 nm.

3.1 Structure :

The X-ray diffraction patterns of indium oxide films deposited as different substrate temperatures are shown in Figure 1. The films deposited at a substrate temperature of 373 K were found to be amorphous in nature while those formed at temperatures ≥ 473 K were polycrystalline and exhibited cubic structure. The films deposited at a substrate temperature of 473 K contained (222), (400) and (440) peaks of indium oxide. When the substrate temperature increased the intensity of (222) peak increased whereas the (400) and (440) peaks intensity decreased. The decrease of the (400) and (440) peak intensity with the increase of substrate temperature indicated that the (222) orientation is more predominant in the films. The lattice parameter evaluated from the (222) peak increased from 1.010 nm to 1.014 nm with the increase of substrate temperature from 473 K to 673 K respectively. The grain size of the films evaluated using the Scherrer's relation [15] increased from 20 nm to 35 nm with the increase of substrate temperature from 473 K to 673 K respectively due to the improvement in the crystallinity of the films.

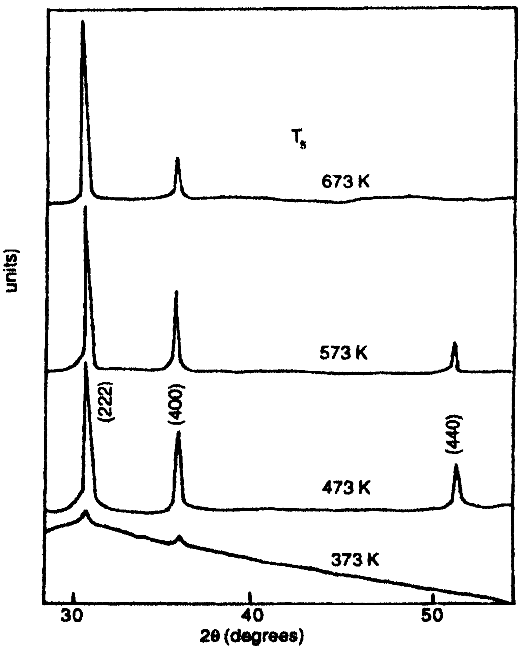

Figure 1. X-ray diffraction patterns of indium oxide films deposited at different substrate temperatures.

3.2 Electrical properties :

The dependence of electrical resistivity (ρ), Hall mobility (μ) and carrier concentration (n) on the substrate temperature of indium oxide films are shown in Figure 2. The electrical resistivity of the films was found to be highly influenced by the substrate temperature. The high electrical resistivity of 2.4×10^{-2} Ω cm in the films formed at 373 K was due to the amorphous nature. The electrical resistivity of the films formed at 473 K decreased to 6 $\times 10^{-3}$ Ω cm due to the polycrystalline nature. The low electrical resistivity of 1.3×10^{-3} Ω cm obtained at a substrate temperature of 673 K was due to the improvement in the degree of crystallinity of the films as revealed by X-ray diffraction data. The Hall mobility measurements indicated that the films were n-type. The Hall mobility of the films increased from 2.5 cm^2/V.sec to 18 cm^2/V.sec and the carrier concentration increased from 1×10^{20} cm^{-3} to 2.7×10^{20} cm^{-3} with the increase of the substrate temperature from 373 K to 673 K respectively. The increase of Hall mobility and carrier concentration with the increase of the substrate temperature might be due to the improvement in the alignment of the grains at the grain boundaries which minimizes the trapping and/or scattering of charge carriers at the grain boundaries. This is a phenomena observed in polycrystalline transparent conducting oxides such as zinc oxide and tin oxide [16]. The data obtained is comparable to the results of Sawada and Higuchi [10], who reported on electrical resistivity of 4.5×10^{-3} Ω cm, Hall mobility of 27 cm/V.sec and carrier concentration of roughly 1×10^{20} cm^{-3} in dc magnetron sputtered films formed by the sputtering of indium target at a substrate temperature of 523 K and an oxygen partial pressure of 1.8×10^{-5} m bar. Golan *et al* [9] reported an electrical resistivity of 4.2×10^{-2} Ω cm on films prepared by sputtering of indium oxide target in pure argon atmosphere and at a substrate temperature of 457 K. Al - Ajili and Bayliss [17] obtained low electrical resistivity of 6×10^{-4} Ω cm on unheated glass substrates by dc reactive magnetron

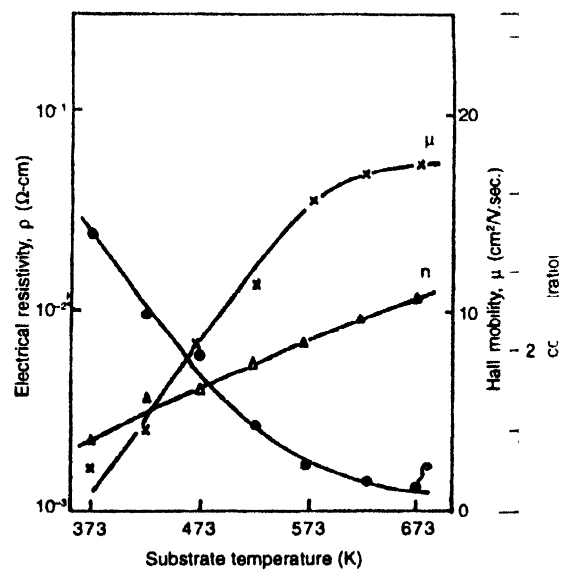

Figure 2. Variation of electrical resistivity (ρ) Hall mobility (μ) and carrier concentration (n) on the substrate temperature of indium oxide films.

sputtering in an oxygen partial pressure of 3.8×10^{-4} mbar. Kasiviswanathan and Rangarajan [8] reported a very high electrical resistivity $10^8\ \Omega$ cm by dc reactive magnetron sputtering on a substrate maintained at room temperature and at an oxygen partial pressure of 2×10^{-3} mbar. The large variation in the electrical resistivities of the indium oxide films formed by the dc magnetron sputtering technique might be due to difference in the deposition parameters employed during the preparation of the films. It is to be noted that the indium oxide films prepared by thermal oxidation of indium, showed electrical resistivity of $8 \times 10^{-3}\ \Omega$ cm when the oxygen / indium ratio was 1.62 [2]. The low electrical resistivity of $2 \times 10^{-4}\ \Omega$ cm obtained in reactive evaporated films [2, 18] was due to the low oxygen / indium ratio of 1.30.

The temperature dependence of Hall mobility was studied in the range 150 – 303 K to understand the electrical transport mechanism operative in indium oxide films. The temperature dependence of Hall mobility was found to obey the Seto's relation [19]

$$\mu = Lq[1/2\pi m^* kT]^{1/2} \exp[-E_b kT], \tag{1}$$

where L is the grain size, q the electron charge, m^* the effective mass of electron, k the Boltzmann constant and E_b the grain boundary potential. Figure 3 shows the plots of $\ln(\mu T^{1/2})$ *versus* $10^3/T$ for the indium oxide films formed at different substrate temperatures. The linearity of the plots indicated that the grain boundary scattering of the charge carriers is more predominant in these films. The grain boundary potential evaluated from these plots decreased from 0.041 eV to 0.025 eV with the increase of substrate temperature from 473 K to 673 K respectively. The decrease of the grain boundary potential with the increase of substrate temperature is due to the increase of grain size. Hence, the decrease of the scattering of electrons at the grain boundaries resulted in the increase of Hall mobility.

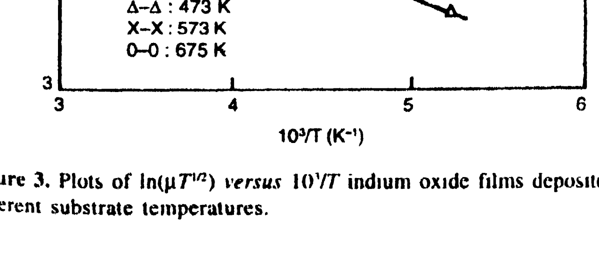

Figure 3. Plots of $\ln(\mu T^{1/2})$ *versus* $10^3/T$ indium oxide films deposited at different substrate temperatures.

3.3 Optical properties :

Figure 4 shows the optical transmittance of the indium oxide films formed at different substrate temperatures. The optical transmittance of the films increased with the increase of substrate temperature. The optical transmittance (at wavelength = 600 nm) of the films increased from 68% to 85% with the increase of substrate temperature from 373 K to 673 K respectively. The optical absorption edge of the films shifted towards lower

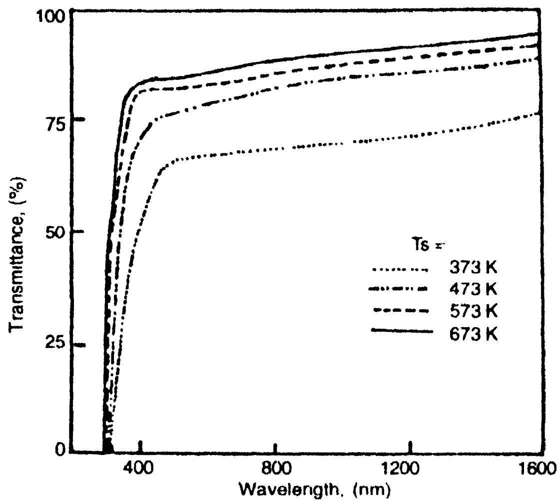

Figure 4. Optical transmittance spectra of indium oxide films deposited at different substrate temperatures

wavelengths at higher substrate temperatures. The optical absorption coefficient (α) of the films was evaluated from the transmittance (T) data by taking into consideration, the reflection losses. The dependence of 'α' on the photon energy ($h\nu$) was found to obey the relation

$$\alpha h\nu = A(h\nu - Eg)^{1/2} \tag{2}$$

where A is a constant and Eg the optical band gap. The plot of $(\alpha h\nu)^2$ *versus* $h\nu$ of indium oxide films formed at different substrate temperatures are shown in Figure 5. The optical band gap of the films was evaluated by extrapolating the linear portion of the plots $(\alpha h\nu)^2$ *versus* $h\nu$ at $\alpha = 0$. The optical band gap of

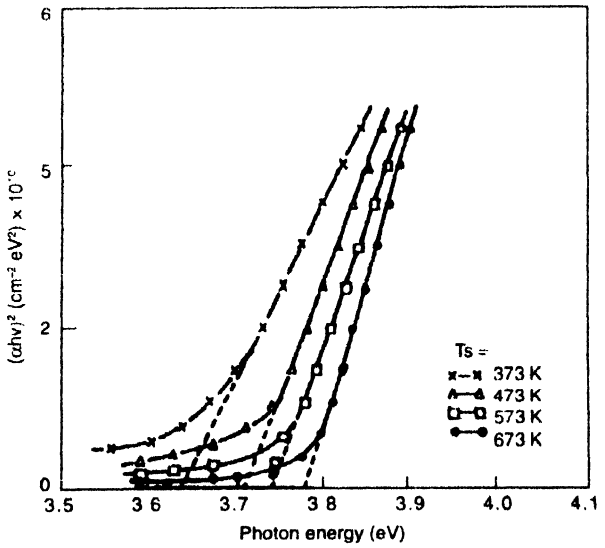

Figure 5. Plots of $(\alpha h\nu)^2$ *versus* $h\nu$ of indium oxide films deposited at different substrate temperatures

the films increased from 3.64 eV to 3.78 eV with the increase of the substrate temperature from 373 K to 673 K respectively. The Hall mobility measurements indicated that the carrier concentration of the films increased with the increase of the substrate temperature. The widening of the optical band gap with the increase of substrate temperature may be attributed to the partial fulfilment of the conduction band by the free carriers due to the blocking of the lower states [20]. Fistul and Vainshtein [21] also noticed that the broadening of the band gap with the increase of the carrier concentration of indium oxide films.

4. Conclusions

Thin films of indium oxide were deposited onto glass substrates maintained at temperatures in the range 373-673 K by employing the dc reactive magnetron sputtering technique. The influence of substrate temperature on the structural, electrical and optical properties were studied. The films formed at 373 K were amorphous in nature whereas those formed at temperatures ≥ 473 K were polycrystalline with cubic structure. The electrical resistivity of the films decreased from 2.4×10^{-2} Ω cm to 1.3×10^{-3} Ω cm and Hall mobility increased from 2.5 cm^2/V.sec to 18 cm^2/V.sec with the increase of substrate temperature from 373 K to 673 K due to the improvement in the crystallinity of the films. The optical transmittance increased from 68% to 85% and the optical band gap increased from 3.64 eV to 3.78 eV with the increase of substrate temperature from 373 K to 673 K respectively.

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